

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Application of : Ayala BARAK  
Appl. No.: 10/586,349 : Group Art Unit: 1616  
Filed : April 10, 2007 : Examiner: Nathan Schlientz  
Confirmation No. : 2789 :  
For : BIOCIDES AND APPARATUS

DECLARATION OF AYALA BARAK UNDER 37 C.F.R. §1.132

I, the undersigned, Dr. Ayala Barak, of 11 Be'eri Street, Tel Aviv, Israel, hereby declare as follows:

1. I am the Applicant in U.S. Patent Application No. 10/586,349 (hereinafter "the application").
2. I have been employed as a professional in the field of disinfection of industrial water. I am currently employed as CEO at A.Y. Laboratories, the assignee of the application. I have a Ph.D. degree in chemistry from the Hebrew University of Jerusalem.
3. In the Office Action dated September 10, 2010, the Examiner states that US 1,590,372 (hereinafter "Harold") discloses producing chloramines from ammonium salts, including ammonium carbamate, and chlorine. The Examiner also states that the presently claimed invention could be arrived at by combining the teachings of Harold and US 5,976,368 (hereinafter "Barak"). As will be shown below, the disclosure in Harold does not teach any aspect of the presently claimed invention.
4. Harold discloses the preparation of germicides, sterilants, etc. and a process of applying them to water, sewage etc. Harold describes the production of "chloramines" in solution at relatively high concentrations. Harold uses the terms

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"chloramine" and dichloramine" as terms of identification only (page 1, lines 14 – 16). Apparently, Harold admits that the product obtained is not necessarily a chloramine by its usual scientific definition. The product is described as having a pungent smell, sharper and more acid than chlorine and a taste in solution similar to chlorine (page 1, lines 16 – 20).

5. Harold describes the method of producing "chloramines" as follows:

- a. The chloramine is produced in relatively high concentrations by forming a mixture of an aqueous solution of chlorine and of an ammonium salt;
- b. The mixture is allowed to stand for a period of 15-45 minutes at room temperature in order for the chlorine to react completely with the ammonium salt;
- c. The mixture which is originally yellow becomes colorless; and
- d. Each molecule of chlorine reacts with 0.1-8 molecules of ammonium salt; the optimum range is 0.28 to 4 molecules of ammonium salt to every molecule of chlorine (page 1, lines 30 – 81).

6. Harold discloses that ammonium carbonate (and its impurities ammonium bicarbonate and ammonium carbamate), ammonium chloride, ammonium nitrate, ammonium sulfate and diammonium hydrogen phosphate are potential ammonium salts for the invention disclosed therein (page 1, line 81– page 2, line 10).

7. The solutions of "chloramine" prepared according to the process disclosed in Harold are preferably employed within a short time after their preparation but if protected from heat and bright sunlight they may be stored for several hours without appreciable decomposition. (page 3, lines 16 – 22).

8. Harold does not describe the pH of the reaction between chlorine and the ammonium salt, nor does it not disclose an optimal pH range. However, the preferred reaction conditions as well as the potential reaction products can be deduced from the disclosure in Harold as follows.

9. Harold uses chlorine gas as the chlorine source for the reactions. Chlorine is a gas that can dissolve in water (0.062 moles/L; Merck Index). Chlorine dissociates in water and forms HOCl and HCl. **Accordingly, the chlorine solution in water is acidic, i.e. the reaction pH is low.**

10. According to Harold, when ammonia forms a salt with a relatively strong acid (e.g. ammonium chloride, ammonium sulfate and ammonium nitrate) the reaction with chlorine proceeds slowly and the resulting "chloramine" remains stable even at high concentration. (page 1, lines 91 – 106). Ammonium salts of strong acids dissolve in water to form an acidic solution, with the pH depending on the concentration and being lower than 6.0. **Thus, the "chloramine" of Harold is produced under acidic conditions.**

11. On the other hand, Harold discloses that ammonium carbonate reacts quickly with chlorine and the resulting "chloramine" formed decomposes as rapidly as it is produced, unless it is formed at low concentration (page 1, lines 82 – 91). Ammonium salts of weak acids (e.g. carbonic acid) dissolve in water to form a neutral to mildly alkaline solution.

12. An article by Weil et al. (J. Am. Chem. Soc. 71(5):1664-1671, 1949), a copy of which is attached hereto (hereinafter "Weil"), describes the chemistry of the reaction between chlorine gas and ammonium ions. Figure 1 on page 1667 shows the dependence of the observed rate of reaction on pH. The reaction is fast at ~pH 8, and it is slower under both acidic and highly alkaline conditions. Weil can be

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used to explain the details of Harold's findings: the reaction between ammonium salts derived from strong acids and chlorine are conducted under acidic conditions, and are thus slower, and can be conducted at high concentrations; the reaction between ammonium carbonate and chlorine occurs at a higher pH (7 – 9, depending on the concentration). The reaction at this higher pH is much faster, and thus cannot be conducted at high concentration.

13. Kumar et al. (Inorg. Chem. 26:3430-3434 (1987), a copy of which is attached herewith (hereinafter "Kumar"), investigated the kinetics and mechanism of the base decomposition of nitrogen trichloride ( $\text{NCl}_3$ ) in aqueous solutions. In the conclusion (p.3434), the authors state that the stability of  $\text{NCl}_3$  in aqueous solutions is greater than previously thought: **" $\text{NCl}_3$  appears to be much less thermodynamically stable than is the case because of its kinetic reactivity with  $\text{NHCl}_2$  in neutral and basic solutions."** This conclusion correlates well with the findings in Harold that when the "chloramine" is produced at low pH (using salts with a strong acid radical) the "chloramine" produced remains stable even at high concentrations, while if the "chloramine" is produced at neutral or higher pH it degrades much faster.

14. All of foregoing indicates that the actual product produced by Harold was nitrogen trichloride, and that it was best produced and stored at low pH. Harold himself indicates that the reactive product may be some "intermediate" rather than "chloramine" or "dichloramine" (page 3, lines 45-61). This conclusion also jibes with Harold's description of the smell and taste of the produced "chloramine".

15. Wen and Brooker (J. Phys. Chem. 99:359-368, 1995), of record (hereinafter "Wen"), presents the equilibria between ammonium carbonate, ammonium bicarbonate, and ammonium carbamate. According to Wen, acidic solutions of

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ammonium carbamate decompose to  $\text{CO}_2$  and  $\text{NH}_4$  (page 362, end of second paragraph.) The carbamate ion ( $\text{H}_2\text{NCOO}^-$ ) is a stable species in neutral and basic aqueous solutions (page 363, last paragraph of left column).

16. If we combine the teachings of Wen, and the understanding that the "chloramine" disclosed by Harold is produced under acidic conditions, it is quite clear that Harold's "chloramines" do not contain ammonium carbamate, and at best contain  $\text{CO}_2$  and/or  $\text{HCO}_3^-$ . The carbamate ion is present only under alkaline conditions.

17. In summary it seems that Harold is producing nitrogen trichloride at high concentrations and low pH. This product is indeed stable as long as the pH is low, and it cannot contain carbamate or ammonium carbamate. In the application, we claim the production of a monochloramine from ammonium carbamate under alkaline conditions. The claimed chloramine is formed immediately as the ingredients are mixed, it is used as soon as it is produced, and it shows better efficacy than a biocide produced from ammonium carbonate (see Example 7).

18. U.S. Patent Nos. 5,976,386 and 6,132,628 (hereinafter "Barak") disclose a method of inhibiting the growth of microorganisms in a liquid comprising adding to the liquid a biocide comprising an oxidant and an amine source. A person of skill in the art, upon reading Harold and Barak, would not think to choose ammonium carbamate, mentioned in Harold, as the amine source for the method of Barak, since the person of skill in the art would understand that the compositions disclosed in Harold could not have contained any ammonium carbamate.

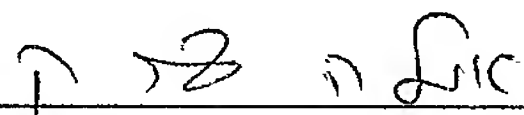
19. A person of skill in the art would also realize that the biocides disclosed in Harold are prepared under acidic conditions, as detailed above, while the biocides



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disclosed in Barak are prepared under alkaline conditions (column 2, lines 42 – 45). Accordingly, a person of skill in the art would not attempt to combine the teachings of Harold and Barak since, at least in terms of pH, the methods disclosed therein are incompatible.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that any such willful false statements may jeopardize the validity of the application or any patent issued thereon.

  
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Ayala Barak, Citizen of Israel  
11 Be'eri Street, Tel Aviv, Israel  
January 3, 2011